



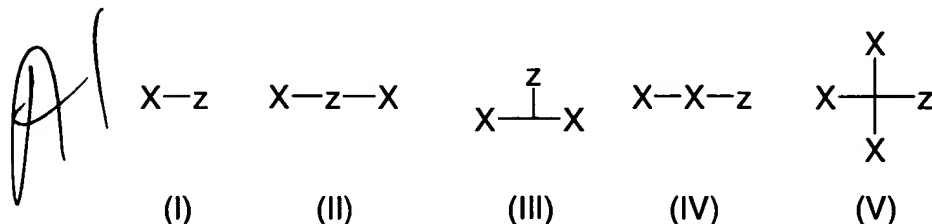
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AMENDMENTS TO THE CLAIMS

TECH CENTER 1600/2000

Claim 1 (Original): A microcapsule comprising an encapsulated material enclosed within a solid permeable shell of a polymer resin wherein the polymer resin has incorporated therein at least one surface modifying compound wherein said surface modifying compound is selected from compounds having a formula (I), (II), (III) (IV) or (V)



wherein Z is a moiety that contributes to modifying the surface properties of said microcapsule and each X is, independently, a functional moiety capable of reacting with a wall-forming material and the moieties designated by lines linking the X and Z functional groups have a molecular weight of between 50 and 4000, and may be optionally substituted aryl, hydrocarbyl, or heterocyclic units, or combinations thereof, optionally containing internally linked amino, ether, thioether, acetal, ester, thioester, amide, sulphonamide, urethane, urea, carbonate, siloxane, or phosphonamide groups or combinations thereof.

Claim 2 (Original): A microcapsule according to claim 1 wherein -X in structures (I) to (III) and (V) is hydroxyl, thiol, a group -NHA wherein A is hydrogen or C<sub>1</sub> to C<sub>4</sub> alkyl or a group -CO-OR where R is hydrogen or a hydrocarbyl moiety having 1-30 carbon atoms optionally linked or substituted by one or more halo, amino, ether or thioether groups or combinations of these or wherein in structure (IV) -X- is -NH-.

Claim 3 (Currently amended): A microcapsule according to claim 1 [or-2] wherein -Z comprises sulphonate, carboxylate, phosphonate, phosphate, quaternary ammonium, betaine, oxyethylene or an oxyethylene-containing polymer.

Claim 4 (Original): A microcapsule according to claim 3 wherein when -Z is sulphonate, carboxylate, phosphonate or phosphate it is present as a salt providing the -Z<sup>-</sup> anion or wherein when -Z is quaternary ammonium it has the structure [-NR<sub>1</sub>R<sub>2</sub>R<sub>3</sub>]<sup>+</sup> A<sup>-</sup>

wherein  $R_1$ ,  $R_2$  and  $R_3$  are independently hydrogen or  $C_1$  to  $C_4$  alkyl and  $A^-$  is a suitable inorganic or organic anion such as halide or acetate provided that not more than one of  $R_1$ ,  $R_2$  and  $R_3$  is hydrogen

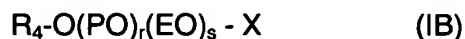
or wherein

when  $-Z$  is oxyethylene or an oxyethylene-containing polymer, it is an oxyethylene polymer or a random or block oxyethylene/oxypropylene copolymer containing an oxyethylene to oxypropylene ratio of greater than 1.

Claim 5 (Currently amended): A microcapsule according to claim 1 ~~[any of the preceding claims]~~ wherein the surface modifying compound of structure (I) has the formula

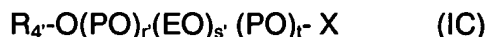


wherein  $Y_1$  represents the moiety linking  $X$  and  $Z$  and is a straight or branched chain alkyl linking group containing from 1 to 20 carbon atoms; or is phenyl, naphthyl, cyclopentyl or cyclohexyl; or or wherein when  $Z$  is an oxyethylene containing polymer and  $Y_1$  represents a direct link between  $X$  and  $Z$  the surface modifying compound of structure (I) has the formula structure (IB)



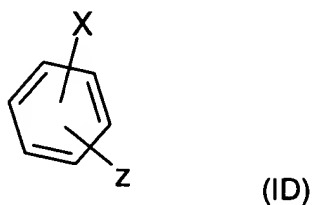
wherein  $R_4$  is an end-capping group which is  $C_1$  to  $C_4$  alkyl,  $r$ , and  $s$  are independently from 0 to 3000, provided that  $s$  is not 0 and the total of  $r + s$  is from about 7 to about 3000 and EO and PO represent oxyethylene and oxypropylene respectively which may be arranged in random or block formation; or

or wherein when  $Z$  is an oxyethylene/oxypropylene block copolymer and  $Y_1$  represents a direct link between  $X$  and  $Z$  the surface modifying compound of structure (I) has the formula (IC)



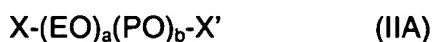
wherein  $R_4'$  is an end-capping group which is  $C_1$  to  $C_4$  alkyl,  $r'$ ,  $s'$  and  $t'$  are independently from 0 to 2000, provided that  $s'$  is not 0 and the total of  $r' + s' + t'$  is from about 7 to about 3000 and EO and PO represent oxyethylene and oxypropylene respectively; or

wherein when  $Y_1$  is a ring structure group, the surface modifying compound of structure (I) has the formula (ID)



wherein X and Z are as defined previously or if X and Z are adjacent substituents capable of reacting together they may form a cyclic anhydride capable or ring-opening under the reaction conditions.

Claim 6 (Currently amended): A microcapsule according to claim 1 [~~any of claims 1 to 4~~] wherein the surface modifying compound of structure (II) wherein -Z- is an oxyethylene containing polymer and there is a direct bond between -Z- and each -X has the formula (IIA) [~~(IIIA)~~]



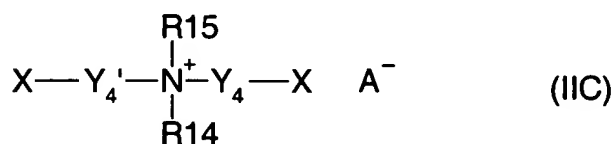
wherein a and b are independently from 0 to 3000, provided that a is not 0 and the total of a + b is from about 7 to about 3000 and EO and PO represent oxyethylene and oxypropylene respectively which may be arranged in random or block formation; or

wherein the surface modifying compound of structure (II) wherein -Z- is an ethylene oxide, propylene oxide block copolymer and there is a direct bond between -Z- and each -X has the formula (IIB)



wherein a', b' and c are independently from 0 to 2000, provided that b is not 0 and the total of a' + b' + c is from about 7 to about 3000 and EO and PO represent oxyethylene and oxypropylene respectively; or

wherein -Z- in structure (II) is quaternary ammonium and structure (II) has the formula (IIC)



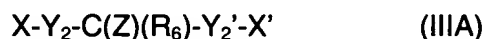
wherein R<sub>14</sub> and R<sub>15</sub>, which may be the same or different, are hydrogen C<sub>1</sub> to C<sub>20</sub> straight or branched chain alkyl; aryl; or C<sub>1</sub> to C<sub>4</sub> aralkyl, wherein each aryl group may be optionally substituted by C<sub>1</sub> to C<sub>4</sub> alkyl, nitro or halo and Y<sub>4</sub> and Y<sub>4</sub>' which may be the same or different are

-R<sub>8</sub>- or

-R<sub>7</sub>-(L<sub>1</sub>)<sub>n</sub>-

wherein R<sub>7</sub>, and R<sub>8</sub> are independently C<sub>1</sub> to C<sub>10</sub> straight or branched chain alkyl linking groups optionally substituted by halogen or C<sub>1</sub> to C<sub>4</sub> alkoxy and (L<sub>1</sub>)<sub>n</sub> is a polyoxyalkylene group; n is from 2 to 20 and A<sup>-</sup> is a suitable anion.

Claim 7 (Currently amended): A microcapsule according to claim 1 [~~any of claims 1 to 4~~] wherein the surface modifying compound of structure (III) has the formula (IIIA)



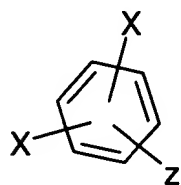
wherein  $R_6$  is hydrogen or more preferably a  $C_1$  to  $C_4$  alkyl group optionally substituted by ether, for example  $C_1$  to  $C_4$  alkoxy or halogen and  $Y_2$  and  $Y_2'$ , which may be same or different are independently

$-R_7-(L_1)_n-$  or

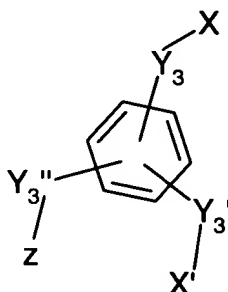
$-R_8-$

wherein  $R_7$ , and  $R_8$  are independently  $C_1$  to  $C_{10}$  straight or branched chain alkyl linking groups optionally substituted by halogen or  $C_1$  to  $C_4$  alkoxy and  $(L_1)_n$  is polyoxyethylene, polyoxypropylene or polyoxybutylene;  $n$  is from 2 to 20 [~~preferably from 4 to 10~~]; or

wherein the surface modifying compound of structure (III) wherein the moiety linking  $X$  and  $Z$  is a ring structure group has the formula (IIIB) or (IIIC)



(IIIB)



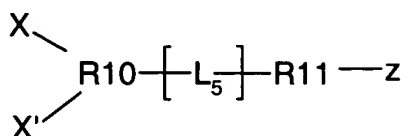
IIIC

wherein  $Y_3$  represents a direct link between  $X$  and the ring structure,  $Y_3'$  represents a direct link between  $X'$  and the ring structure and  $Y_3''$  represents a direct link between  $Z$  and the ring structure or  $Y_3$ ,  $Y_3'$  and  $Y_3''$  individually may be a group [wherein  $Y_3$ ,  $Y_3'$  and  $Y_3''$  individually represent a direct link between  $X$  or  $Z$  (as the case may be) and the ring structure or may be a group]

$-(L_2)-R_9$

where  $L_2$  is an ester linking group  $-C(O)-O-$ ,  $R_9$  is oxyethylene, oxypropylene or oxybutylene or polyoxyethylene, polyoxypropylene or polyoxybutylene having a degree of polymerisation from 2 to 20; or

wherein the surface modifying compound of structure (III) has the formula (IIID)



IIID

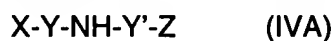
wherein  $R_{10}$  is a  $C_1$  to  $C_8$  straight or branched chain alkyl group and the two groups X and X', which may be the same or different, may be attached to the same carbon atom in the alkyl chain or to different carbon atoms in the alkyl chain,  $-L_5-$  a linking group which is

$-(L_1)_n-$  or

$-R_8-$

wherein  $R_8$ , and  $(L_1)_n$  are as defined above in relation to formula IIIA and  $R_{11}$  is  $C_1$  to  $C_4$  alkyl.

Claim 8 (Currently amended): A microcapsule according to claim 1 [~~any of claims 1 to 4~~] wherein the surface modifying compound of structure (IV) has the formula (IVA)



wherein Y and Y' include independently a straight or branched chain  $C_1$  to  $C_{10}$  alkyl group, a polyoxyethylene, polyoxypropylene or polyoxybutylene polymer chain of formula  $-(L_1)_n-$  as defined above or a group  $-(L_2)-R_9-$  as defined above.

Claim 9 (Currently amended): A microcapsule according to claim 1 [any of the claims 1 to 4] wherein the surface modifying compound is a sulfonate polyester polyol prepared by reacting sodium sulfoisophthalic acid, adipic acid, cyclohexane dimethanol, methoxy-polyethylene glycol (mw 750) and trimethylol propane to give a product having a hydroxyl number in the range of from 150 to 170.

Claim 10 (Currently amended): A microcapsule according to claim 1 [~~any of the preceding claims~~] wherein the solid permeable shell of polymer resin is made by isocyanate polymerisation and the surface modifying compound reacts with the isocyanate moiety in the wall forming material.

Claim 11 (Original): A microcapsule according to claim 10 wherein the isocyanate wall forming material is tolylene diisocyanate or an isomer thereof, phenylene diisocyanate or an isomer thereof, biphenylene diisocyanate or an isomer thereof, polymethylenepolyphenyleneisocyanate (PMPPI), hexamethylene diisocyanate (HMDI) or a trimer thereof or isophoronediiisocyanate (IPDI).

Claim 12 (Currently amended): A microcapsule according to claim 10 [~~or 11~~] wherein the proportions of the surface modifying chemical relative to the wall forming material are such that there is an excess of total isocyanate groups present in the wall forming material over total groups – X.

Claim 13 (Original): A microcapsule according to claim 12 wherein ratio of the total moiety(ies) – NCO in the wall-forming material to the total reactive moiety(ies) –X in the surface modifying compound is from 2 : 1 to 25 : 1

Claim 14 (Currently amended): A microcapsule according to claim 1 [~~any of claims 1 to 9~~] wherein the solid permeable shell of polymer resin is made by the polymerisation of a urea formaldehyde prepolymer in which the methanol (-CH<sub>2</sub>OH) groups have optionally been partially etherified by reaction with a C<sub>4</sub>-C<sub>10</sub> alkanol, and the surface modifying compound reacts with the methanol or etherified methanol moieties in the urea formaldehyde wall forming material.

Claim 15 (Original): A microcapsule according to claim 14 wherein the mole ratio of the surface modifying agent to the number of urea-formaldehyde repeat units in the urea formaldehyde prepolymer is between 1:40 to 1:4.


Claim 16 (Original): A modified process for the encapsulation of a dispersed material within a solid permeable shell of a polymer resin formed by polymerisation of a wall-forming material which comprises incorporating a surface modifying compound having a formula (I), (II), (III), (IV) or (V) as defined in Claim 1 into the polymer resin.

Claim 17 (Original): A process according to claim 16 comprising

- a) reacting the surface-modifying compound with at least one wall-forming material thereby forming a modified surface-active intermediate;
- b) preparing an organic solution or oil phase comprising the material to be encapsulated, the modified surface-active intermediate, and, optionally, additional wall-forming material;
- c) creating an emulsion of the organic solution in a continuous phase aqueous solution comprising water and, optionally, a protective colloid, wherein the emulsion comprises discrete droplets of the organic solution dispersed throughout the continuous phase aqueous solution, with an interface formed between the discrete droplets of organic solution and the aqueous solution; and either
- d) causing *in situ* polymerization and/or curing of the modified wall-forming material in the organic solution of the discrete droplets at the interface with the aqueous solution by heating the emulsion for a sufficient period of time and optionally adjusting the pH to a suitable value to allow substantial completion of wall formation, thereby converting the organic solution droplets to capsules consisting of solid, permeable, polymer shells enclosing the material and having the surface modifying compound incorporated therein, or as an alternative to (d)

e) causing polymerization at the oil-water interface by bringing together a wall forming material added through the aqueous continuous phase and capable of reacting with the wall forming material(s) in the discontinuous oil phase.

Claim 18 (Original): A process according to claim 16 comprising

- 
- a) preparing an organic solution or oil phase comprising the material to be encapsulated, the surface modifying compound and the wall-forming material
  - b) creating an emulsion of the organic solution in a continuous phase aqueous solution comprising water and, optionally, a protective colloid, wherein the emulsion comprises discrete droplets of the organic solution dispersed throughout the continuous phase aqueous solution, with an interface formed between the discrete droplets of organic solution and the aqueous solution; and either
  - c) causing *in situ* polymerization and/or curing of the modified wall-forming material in the organic solution of the discrete droplets at the interface with the aqueous solution by heating the emulsion for a sufficient period of time and optionally adjusting the pH to a suitable value to allow substantial completion of wall formation, thereby converting the organic solution droplets to capsules consisting of solid, permeable, modified polymer shells enclosing the material; or as an alternative to (c)
  - (d) causing polymerization at the oil-water interface by bringing together a wall forming material added through the aqueous continuous phase and capable of reacting with the wall forming material(s) in the discontinuous oil phase.

Claim 19 (Original): A process according to claim 16 comprising

- a) preparing an organic solution or oil phase comprising the material to be encapsulated and the wall-forming material;
- b) creating an emulsion of the organic solution in a continuous phase aqueous solution comprising water and the surface-modifying compound(s), wherein the emulsion comprises discrete droplets of the organic solution dispersed throughout the continuous phase aqueous solution, with an interface formed between the discrete droplets of organic solution and the aqueous solution; and
- c) causing *in situ* polymerization and/or curing of the wall-forming material so that the surface-modifying molecule(s) is incorporated into the wall by heating the emulsion for a sufficient period of time and optionally adjusting the pH to a suitable value, to allow substantial completion of wall formation, thereby converting the organic solution droplets to capsules consisting of solid, permeable, modified polymer shells enclosing the material.

Claim 20 (Original): A process according to claim 16 comprising

- a) preparing an organic solution or oil phase comprising the material to be encapsulated and a first wall-forming material(s) ;
- b) creating an emulsion of the organic solution in a continuous phase aqueous solution comprising water and the surface-modifying compound(s), wherein the emulsion comprises discrete droplets of the organic solution dispersed throughout the continuous phase aqueous solution, with an interface formed between the discrete droplets of organic solution and the aqueous solution whereupon the surface modifying compounds(s) react at the interface with wall forming material from the organic phase; and
- c) causing polymerization at the oil-water interface by bringing together a second wall forming material added through the aqueous continuous phase and capable of reacting with the first wall forming material(s) in the discontinuous oil phase.

Claim 21 (Currently amended): A process according to claim 16 wherein there is employed a combination of the processes [claimed in claims 17 to 20] selected from at least two of the following:

1) a process comprising

- a) reacting the surface-modifying compound with at least one wall-forming material thereby forming a modified surface-active intermediate;
- b) preparing an organic solution or oil phase comprising the material to be encapsulated, the modified surface-active intermediate, and, optionally, additional wall-forming material;
- c) creating an emulsion of the organic solution in a continuous phase aqueous solution comprising water and, optionally, a protective colloid, wherein the emulsion comprises discrete droplets of the organic solution dispersed throughout the continuous phase aqueous solution, with an interface formed between the discrete droplets of organic solution and the aqueous solution; and either
- d) causing *in situ* polymerization and/or curing of the modified wall-forming material in the organic solution of the discrete droplets at the interface with the aqueous solution by heating the emulsion for a sufficient period of time and optionally adjusting the pH to a suitable value to allow substantial completion of wall formation, thereby converting the organic solution droplets to capsules consisting of solid, permeable, polymer shells enclosing the material and having the surface modifying compound incorporated therein, or as an alternative to (d)
- e) causing polymerization at the oil-water interface by bringing together a wall forming material added through the aqueous continuous phase and capable of reacting with the wall forming material(s) in the discontinuous oil phase;



2) a process comprising

- a) preparing an organic solution or oil phase comprising the material to be encapsulated, the surface modifying compound and the wall-forming material
- b) creating an emulsion of the organic solution in a continuous phase aqueous solution comprising water and, optionally, a protective colloid, wherein the emulsion comprises discrete droplets of the organic solution dispersed throughout the continuous phase aqueous solution, with an interface formed between the discrete droplets of organic solution and the aqueous solution; and either
- c) causing *in situ* polymerization and/or curing of the modified wall-forming material in the organic solution of the discrete droplets at the interface with the aqueous solution by heating the emulsion for a sufficient period of time and optionally adjusting the pH to a suitable value to allow substantial completion of wall formation, thereby converting the organic solution droplets to capsules consisting of solid, permeable, modified polymer shells enclosing the material; or as an alternative to (c)
- (d) causing polymerization at the oil-water interface by bringing together a wall forming material added through the aqueous continuous phase and capable of reacting with the wall forming material(s) in the discontinuous oil phase;

3) a process comprising

- a) preparing an organic solution or oil phase comprising the material to be encapsulated and the wall-forming material;
- b) creating an emulsion of the organic solution in a continuous phase aqueous solution comprising water and the surface-modifying compound(s), wherein the emulsion comprises discrete droplets of the organic solution dispersed throughout the continuous phase aqueous solution, with an interface formed between the discrete droplets of organic solution and the aqueous solution; and
- c) causing *in situ* polymerization and/or curing of the wall-forming material so that the surface-modifying molecule(s) is incorporated into the wall by heating the emulsion for a sufficient period of time and optionally adjusting the pH to a suitable value, to allow substantial completion of wall formation, thereby converting the organic solution droplets to capsules consisting of solid, permeable, modified polymer shells enclosing the material; and

4) a process comprising

- a) preparing an organic solution or oil phase comprising the material to be encapsulated and a first wall-forming material(s) ;
- b) creating an emulsion of the organic solution in a continuous phase aqueous solution comprising water and the surface-modifying compound(s), wherein the emulsion comprises discrete droplets of the organic solution dispersed throughout the continuous phase aqueous solution, with an interface formed between the discrete droplets of organic solution and the aqueous solution whereupon the surface modifying compounds(s) react at the interface with wall forming material from the organic phase; and
- c) causing polymerization at the oil-water interface by bringing together a second wall forming material added through the aqueous continuous phase and capable of reacting with the first wall forming material(s) in the discontinuous oil phase.

Claim 22 (Currently amended): A process according to claim 16 [~~any of claims 16 to 20~~] wherein the wall forming material which reacts with the surface modifying compound is an isocyanate wall forming material.

Claim 23 (Currently amended): A process according to claim 16 [~~any of claims 16 to 20~~] wherein the wall forming material which reacts with the surface modifying compound is a urea formaldehyde prepolymer in which the methylol ( $-\text{CH}_2\text{OH}$ ) groups have optionally been partially etherified by reaction with a  $\text{C}_4\text{-C}_{10}$  alkanol.

Claim 24 (Original): The reaction product of a surface modifying compound having the formula (I), (II), (III), (IV) or (V) as defined in claim 1 and an isocyanate wall forming material or a urea formaldehyde prepolymer in which the methylol ( $-\text{CH}_2\text{OH}$ ) groups have optionally been partially etherified by reaction with a  $\text{C}_4\text{-C}_{10}$  alkanol.

Claim 25 (Currently amended): A microcapsule according to claim 1 [~~any of claims 1 to 14 or a process according to any of claims 15 to 21~~] wherein the encapsulated material is an agrochemical, an ink, a dye, a biologically active material or a pharmaceutical.

Claim 26 (Currently amended): A method for modifying the soil mobility of an agrochemical which comprises encapsulating the agrochemical in a process according to claim 16 [~~any of claims 16 to 23~~] and modifying the surface properties by incorporating a surface modifying compound.

Claim 27 (New): A microcapsule according to claim 7 wherein n is from 4 to 10.